ARTICLE IN PRESS

WATER RESEARCH XXX (2010) 1–10

Available at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/watres

Assessment of arsenic exposure from groundwater and rice in Bengal Delta Region, West Bengal, India

Debashis Chatterjee[a,*], *Dipti Halder*[a,b], *Santanu Majumder*[a], *Ashis Biswas*[a,b], *Bibhash Nath*[c], *Prosun Bhattacharya*[b], *Subhamoy Bhowmick*[a], *Aishwarya Mukherjee-Goswami*[a], *Debasree Saha*[a], *Rasmani Hazra*[a], *Palash B. Maity*[d], *Debankur Chatterjee*[e], *Abhijit Mukherjee*[f], *Jochen Bundschuh*[b,g]

[a] *Department of Chemistry, University of Kalyani, Kalyani 741 235, West Bengal, India*
[b] *KTH-International Groundwater Arsenic Research Group, Department of Land and Water Resources Engineering, Royal Institute of Technology (KTH), Teknikringen 76, Stockholm, SE-10044, Sweden*
[c] *School of Environmental Systems Engineering, The University of Western Australia, Crawley WA 6009, Australia*
[d] *Forensic Science Laboratory, Government of West Bengal, 37/1/2 Belgachia Road, Kolkata 700 037, West Bengal, India*
[e] *Presidency College, 86/1 College Street, Kolkata 700 073, West Bengal, India*
[f] *Alberta Geological Survey, Edmonton, AB T6B 2X3, Canada*
[g] *Institute for Applied Research, Karlsruhe University of Applied Sciences, Moltkestraße 30, D-76133 Karlsruhe, Germany*

ARTICLE INFO

Article history:
Received 8 February 2010
Received in revised form
1 April 2010
Accepted 8 April 2010
Available online xxx

Keywords:
Arsenic exposure
Hydrochemistry
Fe Cycling
Redox chemistry
Rice
Risk assessment

ABSTRACT

Arsenic (As) induced identifiable health outcomes are now spreading across Indian subcontinent with continuous discovery of high As concentrations in groundwater. This study deals with groundwater hydrochemistry vis-à-vis As exposure assessment among rural population in Chakdaha block, West Bengal, India. The water quality survey reveals that 96% of the tubewells exceed WHO guideline value (10 μg/L of As). The groundwaters are generally anoxic (−283 to −22 mV) with circum-neutral pH (6.3 to 7.8). The hydrochemistry is dominated by HCO_3^- (208 to 440 mg/L), Ca^{2+} (79 to 178 mg/L) and Mg^{2+} (17 to 45 mg/L) ions along with high concentrations of As_T (As total, below detection limit to 0.29 mg/L), Fe_T (Fe total, 1.2 to 16 mg/L), and Fe(II) (0.74 to 16 mg/L). The result demonstrates that Fe(II)–Fe(III) cycling is the dominant process for the release of As from aquifer sediments to groundwater (and vice versa), which is mainly controlled by the local biogeochemical conditions. The exposure scenario reveals that the consumption of groundwater and rice are the major pathways of As accumulation in human body, which is explained by the dietary habit of the surveyed population. Finally, regular awareness campaign is essential as part of the management and prevention of health outcomes.

1. Introduction

A growing concern over the incidence of widespread human exposure of arsenic (As) has been seriously noticed during the past three decades globally. The increased exposure of As is generally associated with the incidences of cancer and other public health hazards. The occurrence of As (mostly inorganic forms) in groundwater have been documented in several parts

* *Corresponding author*. Tel.: +91 33 2442 3563; fax: +91 33 2582 8282.
E-mail address: dbchat2001@rediffmail.com (D. Chatterjee).
0043-1354/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.watres.2010.04.007

of the world including countries like USA, EU, Australia and several Latin American countries (Guha Mazumdar et al., 1988; Bhattacharya et al., 1997; Nordstrom, 2002; Chakraborti et al., 2004; Kapaj et al., 2006; Nriagu et al., 2007; Bundschuh et al., 2009; Bundschuh et al. 2010, this volume). However, the situation in Southeast Asia, notably in Bengal Delta Plain (BDP, West Bengal) is critical, since As-enriched groundwater is posing a serious threat to the community drinking water supply and human health (Bhattacharya et al., 2002, 2007; Smedley and Kinniburgh, 2002; Chatterjee et al., 2005; Rahman et al., 2009a).

Arsenic compounds are known human carcinogens and their toxicity to mammals has been reported by several workers (Bates et al., 1992; NRC, 1999; IPCS, 2001; ATSDR, 2007). In the BDP, the level of human exposure of As is unprecedented both in terms of geographical area coverage and incidences of identifiable health outcomes (Bhattacharyya et al., 2003a; DNGM, 2008; Rahman et al., 2009a). The chronic exposure of As from groundwater (exposure level: $\geq$0.05 mg/L; duration: $\geq$6 months) often leads to several types of skin manifestations (e.g. melanosis, leucomelanosis, keratosis, hyperkeratosis, etc.), non-pitting edema, respiratory diseases, gastro-intestinal, liver and cardiovascular problems including cancer (Bates et al., 1992; Hopenhayn-Rich et al., 2000; NRC, 2001; Vather and Concha, 2001; Kapaj et al., 2006). However, several studies reveal that skin lesions are relatively minor health outcome and probably occur due to ingestion of groundwater containing high As concentrations ($\geq$0.05 mg/L) for a prolonged ($\sim$8 years) time period (Kapaj et al., 2006; Smith and Steinmans, 2009).

Recent studies have revealed that human exposure of As has resulted from several pathways such as drinking water, food, beverages, soil, inhalation of dust and atmospheric particulates (Bhattacharyya et al., 2003b; Kapaj et al., 2006; Kar et al., 2006; Nriagu et al., 2007; Nath et al., 2008; Naidu and Bhattacharya, 2009). Arsenic once ingested (mostly as oxyanions), can be metabolized in the human system through various processes and pathways to form a large number of methylated species. Among the metabolites, the methylated metabolite of As [*m*As(III)] has shown to be cytotoxic, more potent inhibitor and stronger promoter of oxidative DNA damage (Chen et al., 1997; Thomas et al., 2001). Skin lesions including pigmentation changes and keratosis are commonly thought to be the hallmark features of As exposure including internal and external cancers, peripheral vascular disease, hypertension and diabetes (Chen et al., 1997; Kapaj et al., 2006; Rahman et al., 2009a). Tseng (2009) observed that the variability in the capacity of As methylation among individuals might exert significant control on human As toxicity. Nutritional status is also an important factor in the methylation capacity of individuals and especially folate has an important role in the methylation and excretion of As.

The toxicity of As generally exerts when it enters into the circulatory system, which largely depends on the amount of ingested As (current as well as lifetime) and exposure duration (prolonged as well as discrete) (Caussy, 2003). Several studies have indicated that the consumption of rice is the primary source of As in non-seafood diet, especially in the tropical region (Roychowdhury et al., 2002; Meharg and Rahman, 2003; Williams et al., 2006; Smith et al., 2006; Torres-Escribano et al., 2008). Similarly, in rural Bengal, rice and rice water (i.e. starch water) are considered as the staple food or food substitute, which are mainly consumed thrice a day, during breakfast, lunch and dinner (Halder, 2007; Rahman et al., 2009b). Global consumption of milled rice is ca. 400 million tons in a year (Ricestat, 2007) which represented ca. 50% of total cereal consumption. However, the quality of rice varies widely, depending on the source (e.g. varieties, growing condition and use of fertilizers) and processing techniques (Raab et al., 2009). Therefore, the use of As-enriched groundwater from shallow irrigation wells for cultivation (mostly summer paddy) and food preparation may lead to significant As exposure among the individuals. (Naidu and Bhattacharya, 2009; Rahman et al., 2009b).

Recently, West Bengal State Government, non-governmental organizations (NGO's) and other local agencies have taken up the responsibility to monitor drinking water quality and campaign for As awareness in the rural Bengal. However, major part of the As-enriched region is uncovered under this surveillance program. We therefore have taken an initiative to study whether the people in uncovered part are aware of the As-induced calamities. This study aimed to understand the hydrochemistry of the As-enriched groundwaters that are often used by the rural villagers for domestic (e.g. cooking, drinking, cleaning, etc.) and agricultural purposes. Using questionnaire based field survey, attempts have also been made to evaluate the role of confounding factors (e.g. awareness for health, hygiene, etc.) linking As exposure to human. We expect that the outcome of this investigation may lead to the formulation of revised policy option for the people at risk.

2. Materials and methods

2.1. *The study area*

The study area (Chakdaha block; latitude: 23°00′20″N–23°05′20″N; longitude: 88°31′40″E–88°49′00″E) is an integral part of the world's largest delta (Ganges–Brahmaputra–Meghna deltaic alluvium, GBM system), located in the eastern bank of the river Hooghly, the major distributory of river Ganges (Fig. 1). The study area is adorned with several geomorphological features such as series of meander scars of varied wavelength and amplitude, abandoned channels, oxbow lakes, inter-distributory levees and flood basins with gradual southern slopes (Nath et al., 2005). The land use pattern of the study area is consisting of agricultural land ($\sim$70–75%) where rice cultivation is the major agricultural practice (Jana, 2003; Chatterjee et al., 2003; Mukherjee et al., 2007). The climate is typically hot and humid (temperature ranges between 6 and 43°C, average relative humidity $>$ 65%) with annual rainfall ranges between 1172 and 1635 mm (mean: 1436 mm) (DSH, 2005). The climatic data was recorded between 1999 and 2003.

In the study area, most of the tubewells ($>$ 90%) are privately owned (10–50 m depth), which are generally not considered for monitoring program under the surveillance of drinking water quality campaign organized by the state

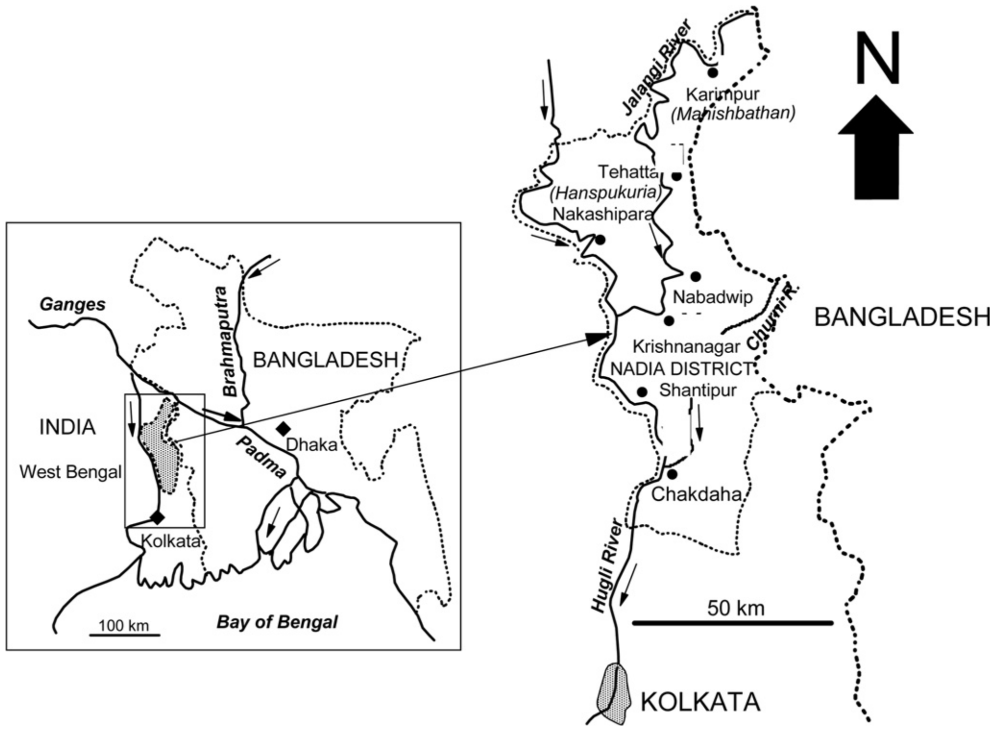

Fig. 1 – Map of the study area in the Bengal Delta Region located at the Chakdaha block, West Bengal, India, an integral part of the Ganges–Brahmaputra–Meghna (GBM) delta showing location of the sampling sites.

government/NGO's/local agencies. There are also a few public tubewells (> 100 m depth) that are commonly used for the community water supply. Large number of irrigation well (~10–50 m depth) are also used mostly during summer months (May–July) for agricultural purpose.

2.2. Sampling and analytical technique

The groundwater samples ($n = 30$) have been collected from the tubewells in pre-cleaned plastic bottles. The samples have been filtered on-site using 0.45 μm membrane filter and divided into two groups. The first group has been acidified with a nitric acid (0.25% v/v, suprapur, MERCK) for the analysis of total As (As_T) and total Fe (Fe_T) using HG/GF mode Atomic Absorption Spectrophotometer (AAS-240, Varian Inc.), while major cation analysis has been performed with the help of Ion Chromatography (761 Compact IC, Metrohm). The second group has been left un-acidified for the analysis of major anions using IC (761 Compact IC, Metrohm). Few field parameters, e.g., pH, Eh, electrical conductivity, temperature, dissolved oxygen and alkalinity (titrating with 0.02 M H_2SO_4) have been measured at the well head. Fe(II) concentrations were measured colorimetrically in the field following the o-phenanthroline method (Jeffrey et al., 1989) using Perkin Elmer Lambda-20 spectrophotometer.

Raw rice samples ($n = 30$) have been collected from each household where groundwater samples have been collected previously. Initially, rice samples have been washed with deionized water (~18 MΩ) and oven dried at ~65 °C for 48 h. Dried rice samples have been grinded manually using a pestle and a mortar to homogenize the material. The homogenized sub-samples (~0.2 g) have been weighted into Pyrex glass digestion tube followed by addition of 2 mL of concentrated HNO_3 (suprapur, MERCK). The homogenized solution has been kept for overnight at a room temperature. The samples were then heated to 120 °C on a heating bath until a clear solution was obtained. The digested samples were then cooled and volume was adjusted to 10 mL. The samples have been analyzed for As_T by using AAS, HG/GF mode (AAS-240, Varian Inc). Rice samples (high aroma long grain and Kohinoor Indian Basmati) collected from the local market have been analyzed in the same manner and used as a control specimen. Commercially available reference materials were also used to ascertain analytical accuracy, which provides quality control of ±10%.

The societal evaluation (age and gender distribution, ethnicity, education and occupation) has been done during water quality survey and at least one principal user of each tubewell has been interviewed in local language (Bengali) using a standardized questionnaire. Awareness level for health, hygiene and As toxicity has been assessed for the study population. Body mass index (BMI, kg/m^2) was also calculated for each of the interviewed users, which has been calculated as the individual's body weight divided by the square of the height of individuals.

3. Results and discussion

3.1. Groundwater hydrochemistry

The survey of the tubewells have been conducted during March 2006 (Phase-I) and 2007 (Phase-II) in the localities of Chakdaha block to demonstrate the groundwater quality

scenarios. The surveyed localities (mouzas, the lowest administrative unit) were not covered under the As awareness campaign and water quality surveillance program recently taken up by the State Government and other agencies. The summary of the physico-chemical parameters of the groundwater in each of the surveyed mouzas is presented in Table 1.

The pH of the groundwater is circum-neutral (6.3–7.8; mean: 7.0) and contains moderate to high electrical conductivity (580–1220 μS/cm; mean: 814 μS/cm). The Eh values (−283 to −22 mV; mean: −105 mV) demonstrate anoxic nature of the aquifer. Among the characteristic chemical features, the groundwater shows high values of alkalinity (208–440 mg/L; mean: 329 mg/L), Ca^{2+} (79–178 mg/L; mean: 113 mg/L), Na^{+} (10–35 mg/L; mean: 20 mg/L), Mg^{2+} (17–45 mg/L; mean: 28 mg/L), SO_4^{2-} (0.96–19 mg/L; mean: 5.7 mg/L), Cl^{-} (8.2–123 mg/L; mean: 35 mg/L), PO_4^{3-} (1.7–7.9 mg/L; mean: 3.6 mg/L) and NO_3^{-} (0.02–4.6 mg/L; mean: 0.37 mg/L).

The groundwater also contains high concentrations of redox sensitive elements such as As_T (below detection limit (bdl) to 0.29 mg/L; mean: 0.10 mg/L), Fe_T (1.2–16 mg/L; mean: 6.4 mg/L) and Fe(II) (0.74–16 mg/L; mean: 6.0 mg/L). It is important to note that only one well (~63 m depth) indicated As concentrations below WHO recommended values (0.01 mg/L). In fact, the survey reveals that groundwater is often exceeding the WHO guideline value as well as the national limit of the Bureau of Indian Standards (BIS, 0.05 mg/L) where no alternative sources are available. The survey of the tubewells shows that 33% of the groundwater ($n = 10$) contains As_T concentration < 0.05 mg/L, whereas another 33% of the wells ($n = 10$) contain As_T between 0.05 to 0.10 mg/L, while the remaining 33% contain $As_T > 0.10$ mg/L. This study also depicts that As concentration in groundwater is spatially variable. The reason could be due to variation in the depositional behavior of the source sediments.

3.2. *Relationship between arsenic, iron and major solutes*

The relationship between As_T and Cl^{-} is moderate (Fig. 2a), which suggests different sources of Cl^{-} in groundwater, e.g., high Cl^{-} is most likely to originate from septic tank leachate, water softening and other household activities. During field survey it has been observed that household sanitations (pit latrine) are coupled with drinking water sources (i.e. tubewells), which might lead to the Cl^{-} enrichment in groundwater. Poor sanitation was also observed throughout rural Bengal where near-surface aquifers were generally enriched with As_T and Cl^{-} (Nath et al., 2008). However, in some cases high As_T tubewells were observed to contain low Cl^{-}.

The relation between Fe_T and HCO_3^{-} is moderate (Fig. 2b), which is also true for As_T and HCO_3^{-} (Fig. 2c). High concentration of Fe_T in groundwater is possibly due to reductive dissolution of Fe-oxyhydroxide solids (i.e. transformation of Fe-III to Fe-II), whereas HCO_3^{-} concentration is mainly induced by the local biogeochemical processes (Bhattacharya et al., 2002; Nath et al., 2008). Fredrickson et al. (1998) demonstrated that the bicarbonate-buffered medium can promote microbial reduction of hydrous ferric oxide because bicarbonate can able to complex with bioproduced Fe(II), and precipitate as siderite, effectively acting as a sink for Fe(II) and As_T removal. Moreover, the cyclic behavior of Fe in the aquifer can also be maintained by the presence of electron donor such as NO_3^{-} (mostly from anthropogenic sources):

1. Present situation (natural)

$$2Fe(OH)_3 + CH_2O + 2H^{+} \rightarrow 2Fe^{2+} + CO_2 + 5H_2O$$

2. Denitrification (natural)

$$4NO_3^{-} + 5CH_2O \rightarrow 2\ N_2\ (g) + 4HCO_3^{-} + CO_2 + 3H_2O$$

3. Nitrate reduction by ferrous ion (anthropogenic)

$$NO_3^{-} + 5Fe^{2+} + 12H_2O \rightarrow 1/2N_2\ (g) + 5Fe(OH)_3 + 9H^{+}$$

This study further demonstrates that the high As tubewells (> 0.1 mg/L) are mostly installed recently (Fig. 2d). The reason for this behavior might be due to discrete carbon source that lowers the redox status of the aquifer. In BDP, millions of tubewells have been drilled using indigenous technique where cow-dung (carbon source) has been used as a local cementing material to reduce the cost of the drilling. The cow-dung is an easily degradable natural organic material (quickly decomposed by the bacteria) which may supply additional carbon to the system and thereby maintaining reducing condition at depth. However, few relatively shallow old tubewells (< 14 m, 12–14 yrs age) with high As concentrations were also observed, which might be as a consequence of continuous source of natural organic matter.

The relation between As_T and SO_4^{2-} (Fig. 2e) is always interesting to note in the BDP due to strong reducing character of the aquifer. The tendency of sulfate reduction in the anoxic environment is important when groundwater contains elevated levels of As and Fe (Nickson et al., 2000; Chatterjee et al., 2003). In the study area, high As and SO_4^{2-} concentrations have been observed, which is possible when the aquifer is relatively near-surface (e.g. < 15 m depth). The household private tubewells located near the pit latrine and/or low-lying areas explains such situation. While, low SO_4^{2-} concentrations in the aquifer suggest occurrence of bacterial sulfate reduction, forming biogenic sulfides (e.g. pyrite).

The depth profile of As shows evidence of As enrichment in the deeper Holocene aquifer of the study area (> 80 m, Fig. 2f). Arsenic in the deeper aquifer is a serious issue because deeper aquifers are regularly used by the villagers for drinking purpose to combat As menace. High As in the deeper aquifer is a new phenomenon in rural Bengal. This suggests that the terminal Pleistocene–Holocene/Pleistocene depositional sequences may have reduced, which has earlier reported to be oxidized and As-free (Smedley and Kinniburgh, 2002). During field drilling campaign, it has observed that such Pleistocene sediments are greenish-brown or olive green in color with moderately reducing character (McArthur et al., 2004). However, these sediments are distinctly different from the oxidized sediments reported from many parts of the Bengal Delta characterized by typical reddish brown/orange color (von Brömssen et al., 2007, 2008; Pal and Mukherjee, 2009).

Please cite this article in press as: Chatterjee, D., et al., Assessment of arsenic exposure from groundwater and rice in Bengal Delta Region, West Bengal, India, Water Research (2010), doi:10.1016/j.watres.2010.04.007

Table 1 – Summary of the salient groundwater chemical parameters and As in the rice collected from the study area.

Mouza-ID	Mouza name	Depth (m)	Age (yr)	pH	Eh (mV)	EC (µs/cm)	Alkalinity (mg/L)	PO_4^{3-} (mg/L)	NO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	Na^+ (mg/L)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Fe_T (mg/L)	Fe(II) (mg/L)	As_T in ground water (mg/L)	As in rice (mg/kg, dw)
I	Ballavpur	95	4	7.4	−88	750	294	5.5	0.09	12	7.2	28	119	32	7.5	6.7	0.04	0.14
	Ballavpur	28	2	7.5	−196	600	250	5.2	0.04	10	6.9	23	99	29	8.8	8.5	0.05	0.12
	Ballavpur	21	14	6.8	−133	900	402	5.0	0.07	27	2.3	27	118	18	8.5	7.6	0.05	0.08
	Ballavpur	14	3	7.3	−40	900	371	5.4	1.2	22	1.6	19	101	29	1.8	1.3	0.05	0.08
II	Mahanala	34	4	6.9	−61	850	395	3.6	0.10	19	12	16	121	26	7.4	6.6	0.10	0.14
	Mahanala	25	4	7.4	−56	1170	440	3.2	0.12	32	8.7	26	109	22	2.5	2.0	0.14	0.12
	Mahanala	37	12	7.1	−50	1220	417	1.7	0.11	54	1.3	23	101	31	2.3	1.9	0.22	0.04
	Mahanala	52	3	7.1	−30	900	252	4.2	0.85	14	4.1	24	114	24	1.6	1.2	0.07	0.06
III	Belghoria	19	3	6.3	−22	920	288	7.2	1.1	35	7.1	15	96	27	2.4	1.7	0.02	0.14
	Belghoria	63	27	7.4	−34	800	308	2.5	0.46	18	19	11	103	19	3.1	2.5	0.00	0.18
	Belghoria	52	15	7.8	−109	900	404	1.8	0.08	20	2.4	10	79	45	6.3	6.0	0.04	0.24
	Belghoria	137	1	7.7	−146	1050	408	2.3	0.07	29	1.1	29	120	37	9.2	8.9	0.08	0.08
	Belghoria	15	4	7.2	−56	580	343	2.2	0.33	8.2	2.1	11	122	18	4.1	3.1	0.09	0.12
IV	Srinagar	33	3	7.8	−283	840	318	2.3	0.03	15	2.0	11	86	30	2.7	2.6	0.05	0.04
	Srinagar	52	2	7.3	−45	730	236	7.9	0.52	11	2.2	11	84	26	3.9	2.9	0.08	0.06
	Srinagar	30	4	7.0	−94	650	304	7.8	0.07	17	3.9	19	104	26	8.4	8.1	0.19	0.06
	Srinagar	30	6	6.8	−142	698	350	4.5	0.06	93	0.98	34	178	40	6.8	6.6	0.13	0.21
	Srinagar	12	1	6.4	−135	690	355	4.3	0.06	15	0.96	12	97	30	14	13	0.29	0.2
V	Purbabishnupur	26	8	7.0	−125	860	340	3.2	4.6	123	18	18	116	23	1.2	0.74	0.28	0.04
	Purbabishnupur	25	3	6.6	−82	666	301	2.1	0.06	23	3.7	18	99	23	7.8	7.6	0.11	0.08
	Purbabishnupur	14	2	7.1	−134	780	298	3.2	0.04	29	7.2	20	118	34	8.3	8.1	0.18	0.04
	Purbabishnupur	45	13	6.9	−132	1100	320	2.8	0.06	62	1.0	22	141	43	11	11	0.13	0.36
	Purbabishnupur	49	2	6.7	−125	785	275	1.8	0.07	11	1.1	19	86	20	16	15.8	0.04	0.08
VI	Rukminidanga	20	9	6.8	−79	910	220	2.0	0.08	44	14	13	144	27	8.3	8.0	0.04	0.12
	Rukminidanga	13	5	7.5	−214	655	208	2.2	0.02	109	1.0	16	124	27	10	9.9	0.04	0.1
VI	Rasullapur	112	10	7.2	−59	740	398	2.2	0.56	72	17	17	136	25	4.1	3.1	0.05	0.12
	Rasullapur	32	5	6.7	−126	615	332	2.5	0.08	19	5.9	29	108	30	5.2	4.9	0.10	0.18
	Rasullapur	95	7	6.6	−109	865	358	3.0	0.05	51	11	16	153	17	8.0	7.7	0.06	0.02
VIII	Balia	32	2	6.5	−135	670	320	3.0	0.05	27	1.3	35	115	45	7.2	7.0	0.09	0.12
	Balia	37	4	6.8	−115	632	359	3.2	0.08	24	2.5	21	111	29	4.9	4.7	0.06	0.14
Minimum		12	1	6.3	−283	580	208	1.7	0.02	8.2	0.96	10	79	17	1.2	0.74	0.00	0.02
Maximum		137	27	7.8	−22	1220	440	7.9	4.6	123	19	35	178	45	16	15.8	0.29	0.36
Mean		42	6.1	7.0	−105	814	328.8	3.6	0.37	35	5.7	20	113	28	6.4	5.99	0.10	0.12

Please cite this article in press as: Chatterjee, D., et al., Assessment of arsenic exposure from groundwater and rice in Bengal Delta Region, West Bengal, India, Water Research (2010), doi:10.1016/j.watres.2010.04.007

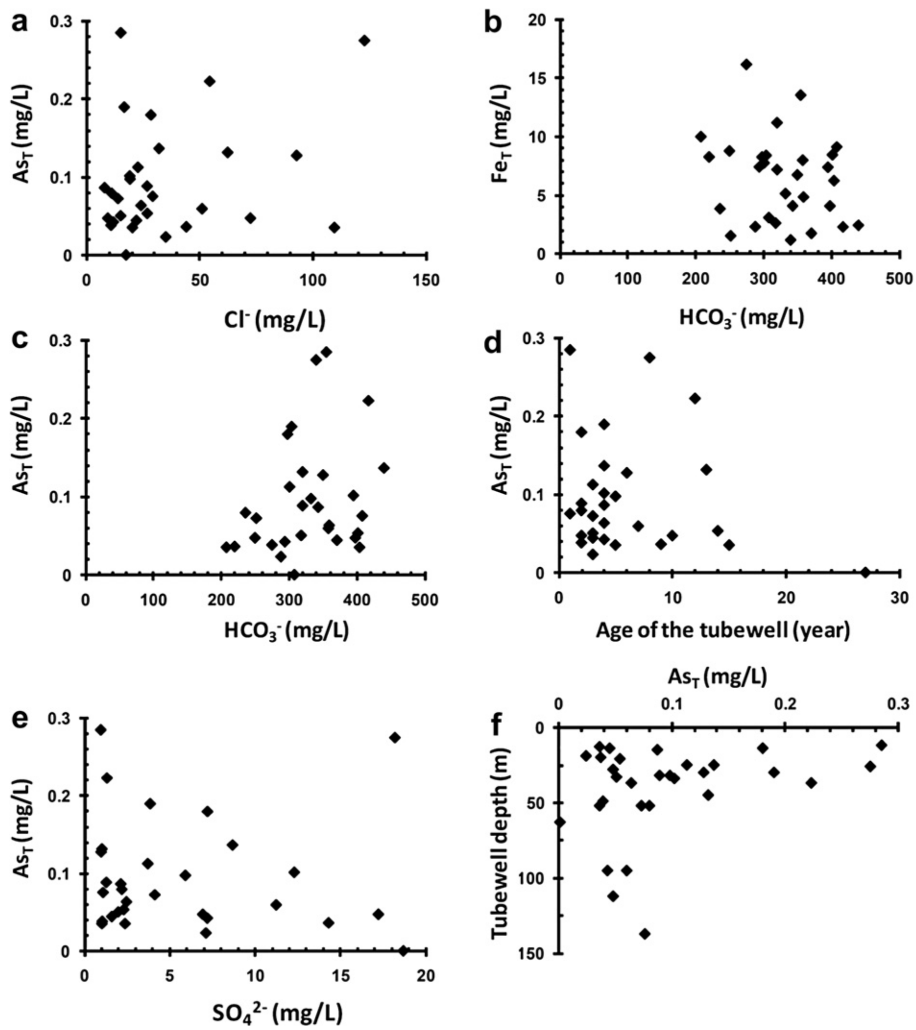

Fig. 2 – Relationship between: (a) As_T and Cl^-; (b) Fe_T and HCO_3^-; (c) As_T and HCO_3^-; (d) As_T and age of the tubewell; (e) As_T and SO_4^{2-}; and (f) depth profile of As_T in groundwater.

3.3. *Arsenic exposure pathway*

It has often observed that the percentage of As patients and their health outcomes are spatially variable and linked with the spatial variation of *i*As in drinking groundwater (e.g. Kapaj et al., 2006). The assessment of *i*As in groundwater and vis-à-vis in rice among surveyed localities (Mouzas) of rural Bengal shows close relationship with each other ($R^2 = 0.47$; Fig. 3). Field experience reveals that such Mouzas generally have wide-spread groundwater pumping from the near-surface aquifer (< 15 m depth) containing elevated As_T concentrations (> 0.3 mg/L) (Halder, 2007). This might result in the accumulation of As in the surface soils, which is transferred to the crops that are grown on these soils and finally transferred to the human system. Previous studies in rural Bengal have shown that accumulation of As in the surface soils was associated with high As concentrations in the underlying aquifer (Nath et al., 2008).

The mean *i*As concentration in the rice samples collected from the study area is 0.12 mg/kg, dry weight (dw) which is comparable with the previous study (0.13 mg/kg, dw) (Williams et al., 2007). This suggests that the dietary intake of rice should receive much attention for understanding the As exposure scenario. Rice is by far the largest dietary source (50 to 70% of the total meal) of *i*As for rural populations even where drinking waters do not contain elevated levels of As.

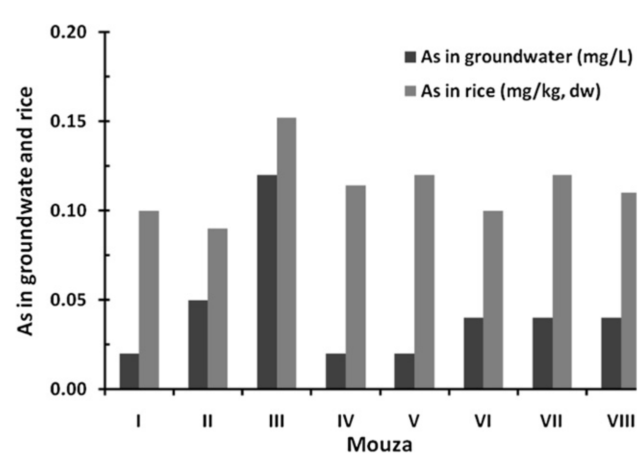

Fig. 3 – Comparison of As_T in groundwater and rice in different mouzas of the study area (Chakdaha block).

Table 2a – Status evaluation (%): age and gender distribution, marital status, ethnicity, education and occupation through Questionnaire Survey.

Age group (yrs)	Gender and marital status (%)			Ethnicity(%)	Education (%)	Occupation (%)
	Gender (%)	Married (%)	Unmarried (%)	Hindu–Muslim		
Subset I (6–10)	Male: 9	0	100	100–0	Illiterate: 12	Farmer: 62
	Female: 8	0	100	100–0	Primary: 55	Shopkeeper: 14
Subset II (11–20)	Male: 11	12.5	87.5	88–12	Secondary: 25	Daily wage labor: 8
	Female: 9	83	17	100–0	Higher secondary: 6	Mason: 3
Subset III (21–30)	Male: 11	80	20	90–10	Undergraduate: 2	Home maker: 10
	Female: 10	100	0	100–0		Others (teachers, health workers): 3
Subset IV (31–40)	Male: 11	100	0	100–0		
	Female: 12	87.5	12.5	100–0		
Subset V (41–50)	Male: 9	100	0	100–0		
	Female: 10	100	0	80–20		

Moreover, the inter-connectivity between different exposure routes (e.g. groundwater and rice) is also an important issue to understand the current role of exposure pathways to human system and its health effects on a spatial scale such as BDP (Khan et al., 2009a,b). The health effects largely depend on the bio-availability of As in the ingested matrices (e.g. drinking water, food, etc.). The ability of *i*As (bio-available forms) absorption into human body followed by entering into circulatory system is the key determinant for the various measures of exposure routes (e.g. water intake, food habits, etc.) and its effect (e.g. skin lesions). However, the exposure and the effect of As to human is mainly depends on the biochemical affinity of different forms of *i*As to human tissues, but, such affinity largely depends on the distribution of As species among the tissues, kinetics of deposition and varying methylation process.

3.4. *Potential confounding factors for arsenic exposure into human system*

This study also focused on the potential confounding factors apart from the As contents in food and water and exposure time. The most important factors are dietary and lifestyle habits, socio-economic status, demographic variables, education and hygiene. A door to door campaign has been conducted during the study period with the help of a well structured set of questionnaire. The influence of the factors has been evaluated against age, ethnicity, gender, marital status, education, occupation, consumption of food and groundwater, awareness levels for health, hygiene and As toxicity, and body mass index (BMI). The outcome of the questionnaire survey is presented in Tables 2a and 2b.

The survey reveals that male (51%) has dominated the study population, where people are literate mostly up to primary level (55%). The major occupation of the survey population is farming (Table 2a). It has also noticed that women have been largely kept in the dark about the threat of As toxicity and in most instances they were the victims of the social taboo and/or stigma. The observed BMI of the individuals could be another important determining criterion for the As ingestion risk assessment (Fig. 4). The scrutiny of the analytical data indicates that the BMI has a closer association with the ingestion risk of As from rice (marked by arrows in Fig. 4) in comparison to ingestion risk of As from groundwater.

Survey of the awareness level for health and hygiene has been made among the population at risk (Fig. 5 and Table 2b). The population at risk has been divided into five age groups such as subset I (0–10 yrs), subset II (11–20 yrs), subset III (21–30 yrs), subset IV (31–40 yrs) and subset V (41–50 yrs). The data shows that awareness level is low among children (subset I) and older-aged (subset V) population. On the other hand, awareness level is relatively high among adolescents, the young adults and the middle-aged working population (i.e. subset II, III and IV). Those age group people may have the opportunity to interact with the society and therefore can exchange their views with others and quickly accumulate information on the identifiable symptoms of arsenicosis as well as management to minimize the risk of As exposure.

Table 2b – Status evaluation of As exposure via drinking water (mg/L) and rice (mg/kg) among various age groups, BMI (kg/m^2) and awareness levels for health, hygiene and As toxicity through Questionnaire Survey.

Age group (yrs)	Arsenic consumption through drinking water (mg/L)	Arsenic consumption through rice (mg/kg, dw)	Awareness levels for health and hygiene (%)	Awareness level for arsenic toxicity (%)	Body Mass Index (BMI) (kg/m^2)
Subset I (6–10)	0.018	0.08	58	50	19
Subset II (11–20)	0.027	0.13	78	78	20
Subset III (21–30)	0.039	0.20	78	93	24
Subset IV (31–40)	0.036	0.18	73	93	22
Subset V (41–50)	0.027	0.20	83	75	22

Please cite this article in press as: Chatterjee, D., et al., Assessment of arsenic exposure from groundwater and rice in Bengal Delta Region, West Bengal, India, Water Research (2010), doi:10.1016/j.watres.2010.04.007

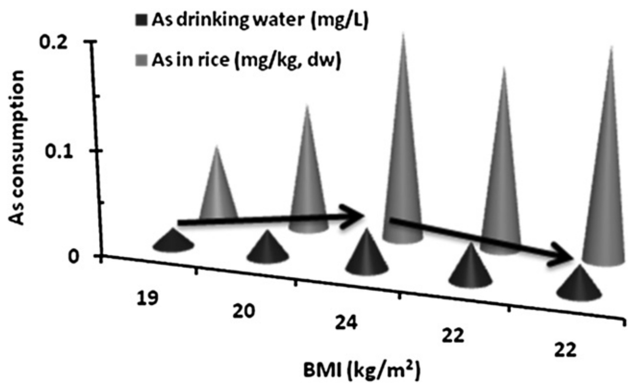

Fig. 4 – Relationship of BMI (kg/m^2) with AS_T consumption from drinking water (mg/L) and rice (mg/kg, dw).

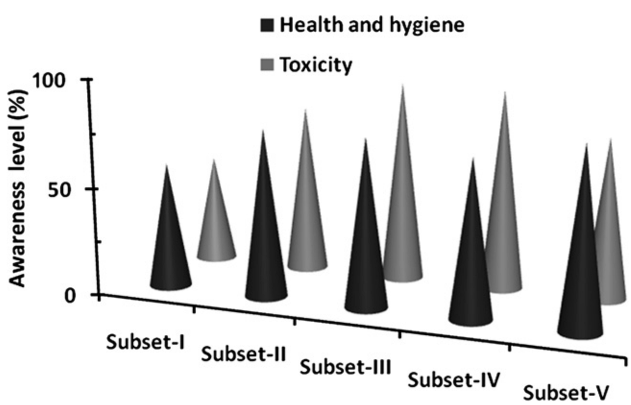

Fig. 5 – Comparison of awareness level for health and hygiene, and As toxicity among different age group population.

4. Conclusion

This study demonstrates that the redox chemistry (in particular reduction of Fe-oxides/hydroxides) and the local biogeochemical conditions regulate the dynamics of *i*As in groundwater. The questionnaire survey reveals that both children and older people have low awareness level for As toxicity compare to adolescents, the young adults and the middle-aged people. The BMI seems to be an important determining criterion for the As ingestion risk among the individuals. The As exposures through various pathways (e.g. drinking groundwater, food and dietary habits) demonstrate urgent need for regulatory measures to optimize the mitigation strategies leading to the formulation of revised policy option for ensuring human health in the affected regions.

Acknowledgements

Corresponding author (DC) is duely acknowledging the long-term collaboration with the Royal Institute of Technology (KTH), Stockholm, Sweden on the investigations of high arsenic groundwaters in the Bengal Delta. We are especially grateful to our collaborators from the USA (Prof. J. Nriagu) for valuable discussions on the health effects of arsenic. Financial support from the University of Kalyani and SAP – UGC program are also duly acknowledged. The authors especially PB thank Sida and the Strategic Environmental Research Foundation (MISTRA) for their financial support through the respective grants, contributions 73000854 and 2005-035-137, respectively during various stages of the present study. Finally, we sincerely thank two anonymous reviewers and the editors for their through and constructive suggestions to give proper shape to the manuscript.

REFERENCES

Agency for Toxic Substances & Disease Registry (ATSDR), 2007. Toxicological Profile for Arsenic. US Department of Health and Human Services, Atlanta, Georgia.

Bates, M.N., Smith, A.H., Hopenhayn-Rich, C., 1992. As ingestion and internal cancers: a review. American Journal of Epidemiology 135, 462–476.

Bhattacharya, P., Chatterjee, D., Jacks, G., 1997. Occurrence of arsenic contaminated groundwater in alluvial aquifers from Delta Plains, Eastern India: options for safe drinking water supply. International Journal of Water Resources Development 13, 79–92.

Bhattacharya, P., Jacks, G., Ahmed, K.M., Khan, A.A., Routh, J., 2002. Arsenic in groundwater of the Bengal Delta Plain aquifers in Bangladesh. Bulletin of Environmental Contamination and Toxicology 69, 538–545.

Bhattacharya, P., Welch, A.H., Stollenwerk, K.G., McLaughlin, M.J., Bundschuh, J., Panaullah, G., 2007. Arsenic in the environment: biology and chemistry. Science of the Total Environment 379, 109–120.

Bhattacharyya, R., Chatterjee, D., Nath, B., Jana, J., Jacks, G., Vahter, M., 2003a. High arsenic groundwater: mobilization, metabolism and mitigation – an overview in the Bengal Delta Plain. Molecular and Cellular Biochemistry 253, 347–355.

Bhattacharyya, R., Jana, J., Nath, B., Sahu, S.J., Chatterjee, D., Jacks, G., 2003b. Groundwater arsenic mobilization in Bengal Delta Plain, the use of ferralite as a possible remedial measure – a case study. Applied Geochemistry 18, 1435–1451.

Bundschuh, J., García, M.E., Birkle, P., Cumbal, L.H., Bhattacharya, P., 2009. Occurrence, health effects and remediation of arsenic in groundwaters of Latin America. In: Bundschuh, J., Armienta, M.A., Birkle, P., Bhattacharya, P., Matschullat, J., Mukherjee, A.B., Bundschuh, J., Bhattacharya, P. (Eds.), Natural Arsenic in Groundwater of Latin America – Occurrence, Health Impact and Remediation. Interdisciplinary Book Series: "Arsenic in the Environment", vol. 1. CRC Press/Balkema, Leiden, The Netherlands, ISBN 978-0-415-40771-7, pp. 3–16.

Bundschuh, J., Litter, M., Ciminelli, V., Morgada, M.E., Cornejo, L., Garrido Hoyos, S., Hoinkis, J., Alarcón-Herrera, M.T., Armienta, M.A., Bhattacharya, P., 2010. Emerging mitigation needs and sustainable options for solving the arsenic problems of rural and isolated urban areas in Latin America - A critical analysis. Water Res. 2010, this volume. doi:10.1016/j.watres.2010.04.001.

Caussy, D., 2003. Case studies of the impact of understanding bioavailability: arsenic. Ecotoxicology and Environmental Safety 56, 164–173.

Chakraborti, D., Sengupta, M.K., Rahman, M.M., Ahamed, S., Chowdhury, U.K., Hossain, M.A., Mukherjee, S.C., Pati, S., Saha, K.C., Dutta, R.N., Zaman, Q.Q., 2004. Groundwater

arsenic contamination and its health effects in the Ganga–Meghna–Brahmaputra plain. Journal of Environmental Monitoring 6, 74–83.
Chatterjee, D., Chakraborty, S., Nath, B., Jana, J., Bhattacharya, R., Basu Mallik, S., Charlet, L., 2003. Mobilization of arsenic in sedimentary aquifer vis-à-vis subsurface iron reduction processes. Journal de Physique IV 1007, 293–296.
Chatterjee, D., Roy, R.K., Basu, B.B., 2005. Riddle of arsenic in groundwater of Bengal Delta Plain – role of non-inland source and redox traps. Environmental Geology 49, 188–206.
Chen, C.J., Chiou, H.Y., Huang, W.I., Chen, S.Y., Hsueh, Y.M., Tseng, C.H., Lin, L.J., Shyu, M.P., Lai, M.S., 1997. Systematic non-carcinogenic effects and developmental toxicity of inorganic arsenic. In: Abernathy, C.O., Calderon, R.L., Chappell, W.R. (Eds.), Arsenic Exposure and Health Effects. Chapman & Hall, London, pp. 124–134.
District Statistical Handbook (DSH), 2005. Statistical database of the Nadia district of West Bengal. Bureau of Applied Economics and Statictics, Govt. of West Bengal, India.
DNGM Research Foundation (DNGM), 2008. Health Effects of Chronic Arsenic Toxicity and Its Mitigation. GTZ-ASEM, German Environmental Programme, New Delhi.
Fredrickson, J.K., Zachara, J.M., Kennedy, D.W., Dong, H., Onstott, T.C., Hinman, N.W., Li, S., 1998. Biogenic iron mineralization accompanying the dissimilatory reduction of hydrous ferric oxide by a groundwater bacterium. Geochimica et Cosmochimica Acta 62, 3239–3257.
Guha Mazumdar, D.N., Chakraborty, A.K., Ghosh, A., Gupta, J.D., Chakraborty, D.P., Dey, S.B., Chattopadhyay, N., 1988. Chronic arsenic toxicity from drinking tubewell water in rural West Bengal. Bulletin of the World Health Organization 66, 499–506.
Halder, D., 2007. Arsenic exposure and health impact. Unpublished Master Dissertation, University of Kalyani, Nadia, West Bengal, India.
Hopenhayn-Rich, C., Browning, S., Hertz-Picciotto, I., Ferreccio, C., Peralta, C., Gibb, H., 2000. Chronic arsenic exposure and risk of infant mortality in two areas in Chile. Environmental Health Perspectives 108 (108), 667–673.
IPCS, 2001. In: Arsenic and Arsenic Compounds, second ed. WHO and WHO-WBVA, Geneva, Switzerland English and Bengali version.
Jana, J., 2003. Genesis of arseniferous groundwater in the BDPs in West Bengal, Eastern India. Doctoral Thesis, University of Kalyani, West Bengal, India. 70 pp. (Unpublished).
Jeffrey, G.H., Bassett, J., Mendham, J., Denney, R.C., 1989. Vogel's Textbook of Quantitative Chemical Analysis. 5th ed. Longman Scientific and Technical, Essex, England. co-published in the United States with John Wiley & Sons, Inc., New York.
Kapaj, S., Peterson, H., Liber, K., Bhattacharya, P., 2006. Human health effects from chronic arsenic poisoning – a review. Journal of Environmental Science and Health, Part A Toxic/Hazardous Substances and Environmental Engineering 41, 2399–2428.
Kar, S., Nath, B., Samal, A.C., Santra, S.C., 2006. Arsenic in urban particulates – a case study in Kolkata Metropolis. Current Science 90, 158–160.
Khan, N.I., Owens, G., Bruce, D., Naidu, R., 2009a. An effective dietary survey framework for the assessment of total dietary arsenic intake in Bangladesh: part-A – FFQ Design. Environmental Geochemistry and Health 31, 207–220.
Khan, N.I., Bruce, D., Naidu, R., Owens, G., 2009b. Implementation of food frequency questionnaire for the assessment of total dietary arsenic intake in Bangladesh: part-B – preliminary findings. Environmental Geochemistry and Health 31, 221–238.
Meharg, A.A., Rahman, M.M., 2003. As contamination of Bangladesh paddy field soils: implications for rice contribution to As consumption. Environmental Science and Technology 37, 229–234.
McArthur, J.M., Banerjee, D.M., Hudson-Edwards, K.A., Mishra, R., Purohit, R., Ravenscroft, P., Cronin, A., Howarth, R.J., Chatterjee, A., Talumder, T., Lowry, D., Houghton, S., Chadha, D.K., 2004. Natural organic matter in sedimentary basins and its relation to As in anoxic ground water: the example of West Bengal and its worldwide implications. Applied Geochemistry 19, 1255–1293.
Mukherjee, P., Chatterjee, D., Jana, J., Maity, P.B., Goswami, A., Saha, H., Sen, M., Nath, B., Shome, D., Sarkar, M.J., Bagchi, D., 2007. Household water treatment option: removal of arsenic in presence of natural Fe-containing groundwater by solar oxidation. In: Bhattacharya, P., Mukherjee, A.B., Bundschuh, J., Zevenhoven, R., Loeppert, R.H. (Eds.), Arsenic in Soil and Groundwater Environment: Biogeochemical Interactions, Health Effects and Remediation. Trace Metals and other Contaminants in the Environment, vol. 9. Elsevier B.V., Amsterdam, The Netherlands, pp. 603–622.
Naidu, R., Bhattacharya, P., 2009. Arsenic in the environment – risks and management strategies. Environmental Geochemistry and Health 31, 1–8.
Nath, B., Berner, Z., Basu Mallik, S., Chatterjee, D., Charlet, L., Stueben, D., 2005. Characterization of aquifers conducting groundwaters with low and high arsenic concentrations: a comparative case study from West Bengal, India. Mineralogical Magazine 69, 841–853.
Nath, B., Sahu, S.J., Jana, J., Mukherjee-Goswami, A., Roy, S., Sarkar, M.J., Chatterjee, D., 2008. Hydrochemistry of arsenic-enriched aquifer from rural West Bengal, India: a study of arsenic exposure and mitigation option. Water Air and Soil Pollution 190, 95–113.
National Research Council (NRC), 1999. Arsenic in Drinking Water. National Academic Press, Washington, DC.
National Research Council (NRC), 2001. Arsenic in Drinking Water. National Academic Press, Washington, DC.
Nickson, R.T., McArthur, J.M., Ravenscroft, P., Burgess, W.G., Ahmed, K.M., 2000. Mechanism of arsenic release to groundwater, Bangladesh and West Bengal. Applied Geochemistry 15, 403–413.
Nordstrom, D.K., 2002. Worldwide occurrences of arsenic in groundwater. Science 296, 2143–2144.
Nriagu, J.O., Bhattacharya, P., Mukherjee, A.B., Bundschuh, J., Zevenhoven, R., Loeppert, R.H., 2007. Arsenic in soil and groundwater: an overview. In: Bhattacharya, P., Mukherjee, A. B., Bundschuh, J., Zevenhoven, R., Loeppert, R.H. (Eds.), Arsenic in Soil and Groundwater Environment: Biogeochemical Interactions, Health Effects and Remediation. Trace Metals and Other Contaminants in the Environment, vol. 9. Elsevier B.V., Amsterdam, The Netherlands, pp. 3–60. doi:10.1016/S0927-5215(06)09001-1.
Pal, T., Mukherjee, P.K., 2009. Study of subsurface geology in locating arsenic-free groundwater in Bengal delta, West Bengal, India. Environmental Geology 56, 1211–1225.
Raab, A., Feldmann, J., Meharg, A.A., 2009. Levels of Arsenic in Rice: The Effects of Cooking, Report #C01049. 27 pp. Available from:. Foods Standard Agency, UK http://www.foodbase.org.uk//admintools/reportdocuments/322–1–599_LEVELS_of_ARSENIC_in_RICE_-_EFFECTS_OF_COOKING.pdf (accessed March 31, 2010).
Rahman, M.M., Naidu, R., Bhattacharya, P., 2009a. Arsenic contamination in groundwater in the Southeast Asia region. Environmental Geochemistry and Health 31, 9–21.
Rahman, M.M., Owens, G., Naidu, R., 2009b. Arsenic levels in rice grain and assessment of daily dietary intake of As from rice in As-contaminated regions of Bangladesh. Environmental Geochemistry and Health 31, 179–187.
Ricestat. Available from:, 2007. International Rice Research Institute (IRRI) http://www.irri.org/science/ricestat/index.asp (accessed March 26, 2010).
Roychowdhury, T., Uchino, T., Tokunaga, H., Ando, M., 2002. Survey of As in food composites from an As-affected area of West Bengal, India. Food and Chemical Toxicology 40, 1611–1621.

Smedley, P.L., Kinniburgh, D.G., 2002. A review of the source, behavior and distribution of arsenic in natural waters. Applied Geochemistry 17, 517–568.
Smith, H.A., Steinmans, M.C., 2009. Health effects of arsenic and chromium in drinking water: recent human findings. Annual Review of Public Health 30, 1–16.
Smith, N.M., Lee, R., Heitkemper, D.T., Cafferky, K.D., Haque, A., Henderson, A.K., 2006. Inorganic arsenic in cooked rice and vegetables from Bangladeshi households. Science of the Total Environment 370, 294–301.
Thomas, D.J., Styblo, M., Lin, S., 2001. The cellular metabolism and systemic toxicity of As. Toxicology and Applied Pharmacology 15, 127–144.
Torres-Escribano, S., Leal, M., Vélez, D., Montoro, R., 2008. Total and inorganic arsenic concentrations in rice sold in Spain, effect of cooking, and risk assessments. Environmental Science and Technology 42 (10), 3867–3872. doi:10.1021/es071516m.
Tseng, C.-H., 2009. A review on environmental factors regulation arsenic methylation in humans. Toxicology and Applied Pharmacology 235, 338–350.
Vather, M., Concha, G., 2001. Role of metabolism in arsenic toxicity. Pharmacology and Toxicology 89, 1–5.
von Brömssen, M., Jakariya, M., Bhattacharya, P., Ahmed, K.M., Hasan, M.A., Sracek, O., Jonsson, L., Lundell, L., Jacks, G., 2007. Targeting low-arsenic in Matlab Upazila, Southeastern Bangladesh. Science of the Total Environment 379 (2–3), 121–132.
von Brömssen, M., Larsson, S.H., Bhattacharya, P., Hasan, M.A., Ahmed, K.M., Jakariya, M., Sikder, A.M., Sracek, O., Bivén, A., Doušová, B., Patriarca, C., Thunvik, R., Jacks, G., 2008. Geochemical characterisation of shallow aquifer sediments of Matlab Upazila, Southeastern Bangladesh – implications for targeting low-As aquifers. Journal of Contaminant Hydrology. 99 (1–4), 137–149.
Williams, P.N., Islam, M.R., Adomako, E.E., Roab, A., Hossian, S.A., Zhu, Y.G., Feldmann, J., Meharg, A.A., 2006. Increase in rice grain arsenic for regions of Bangladesh irrigating paddies with elevated arsenic in groundwater. Environmental Science and Technology 40, 4903–4908.
Williams, P.N., Rabb, A., Feldmann, J., Meharg, A.A., 2007. Market basket rice shows elevated levels of arsenic in South Central U.S. processed rice compared to California: consequences for human dietary exposure. Environmental Science and Technology 41, 2178–2183.

Please cite this article in press as: Chatterjee, D., et al., Assessment of arsenic exposure from groundwater and rice in Bengal Delta Region, West Bengal, India, Water Research (2010), doi:10.1016/j.watres.2010.04.007